

Chemical Reaction Engineering
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Lecture No. # 20
Gas-Solid Catalytic Reactions – Diffusion and Reaction I

Friends, let us continue our discussion on heterogeneous gas solid catalytic reactions. And let us recap what we saw in the last session, namely we set up the mass balances for a porous catalyst having slab geometry and we considered first order irreversible reaction in a isothermal catalyst pellet. As I mentioned we will eventually relax all these restrictions, namely first order reaction, isothermal pellet and slab geometry and look at results for different scenarios.

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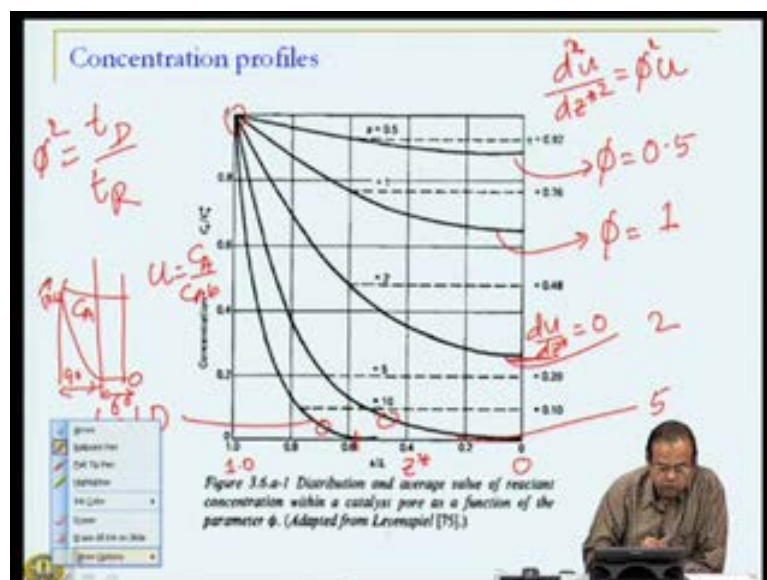
The slide shows a diagram of a catalyst slab of length \$L\$ with a differential element of thickness \$dz\$ at position \$z\$. The bulk phase concentration is \$C_{Ab}\$. Inside the catalyst, concentration \$C_A\$ decreases from \$C_{Ab}\$ at \$z=0\$ to 0 at \$z=L\$. The diagram labels 'Diffusion' and 'Reaction'. To the right, the mass balance terms are listed: Accumulation, In, Out, and Generated. Below the diagram, handwritten equations define the Thiele modulus \$\phi^2 = \frac{L^2 k_{1B}}{D_{eA} C_{Ab}}\$ and the effectiveness factor \$\eta = \frac{\int_0^L C_A dz}{C_{Ab} L}\$. The governing equation is \$\frac{\partial}{\partial t}(\rho_r C_A) = \frac{\partial}{\partial z} \left(D_{eA} \frac{\partial C_A}{\partial z} \right) - r = 0\$. Boundary conditions are given as \$z=0, \frac{dC_A}{dz} = 0\$ and \$z=L, C_A = 0\$.

So let us recap, what we have is a catalyst, which is porous. This is the centre line of the catalyst. So, there is an equivalent catalyst on the right hand side. So, this is our \$z\$ equal to 0. This is our \$Z\$ equal to \$l\$; the gaseous reactant is outside in the bulk. We are right now not considering any external mass transfer resistances. So, the concentration at the surface is \$C_{Ab}\$ and as the reactant travels inside the catalyst, there is diffusion and there is a reaction. The net result of writing a mass balance for this situation is, we get a unsteady state mass balance which looks something like this, term on the left hand side

denoting the accumulation, term the first term on the right hand side denoting the diffusion phenomena, the second term the reaction.

This is an unsteady state diffusion reaction equation, which we simplified saying that at steady state, that means, this term goes to 0 and we have this equal to 0. We also made further simplification saying that we are going to assume or make equations dimensionless. So, we define dimensionless concentration as C_A by $C_{A,b}$. we define dimensionless length Z^* as Z by l and wrote the mass balance equation as $d^2 u / dz^{*2} = -\phi^2 u$. This ϕ^2 , assuming we are assuming first order reaction, so this works out to be. The ratio of $l^2 k v$ and $D A e$ which we also interpreted as the ratio of time scale for diffusion to time scale for reaction. The boundary conditions for this particular equation is at Z^* equal to 0 $dC_A / dz^* = 0$ rather $D u / dz^* = 0$ and at Z^* equal to 1 $u = 1$. So now, let us look at how does this solution of these equation is a second order ordinary differential equation. It is a linear equation.

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So, you can use analytical methods to solve this equation. And you get a solution which looks something like this. What we are looking here is our z^* being 0 here and 1 over here on the other side. And this is our concentration u , which is C_A by $C_{A,b}$, again once again between 0 and 0 and 1. Because of boundary condition that at z^* equal to 1 u is

equal to 1 all solutions start from this point, but what we are seeing in this particular figure is the solution of this equation, let me write the equation once again $d^2 u / dz^2 = \phi^2 u$ with the appropriate boundary condition.

Only parameter what we have is the Thiele modulus ϕ and so, if we change the value of this modulus ϕ which is $\sqrt{k_v / D} \cdot A \cdot e$. Then for different values of ϕ for example, this is a solution for ϕ equal to 0.5, this is a solution for ϕ equal to 1, this 1 for 2, this 1 for 5, this is for ϕ equal to 10. Let us examine this solution, as I said all these solutions start at u equal to 1 and at centre point, du/dz is equal to 0 for all of them, because that is our boundary condition. Our solution has to satisfy this particular equation. Now, let us look what happens when ϕ equal to 0. Remember we defined ϕ^2 , as time scale for diffusion to time scale for reaction. When ϕ is small value, we will call these values point ϕ 1, 2, 5, 10 related to each other. So, when ϕ is 0.5, which is relatively low.

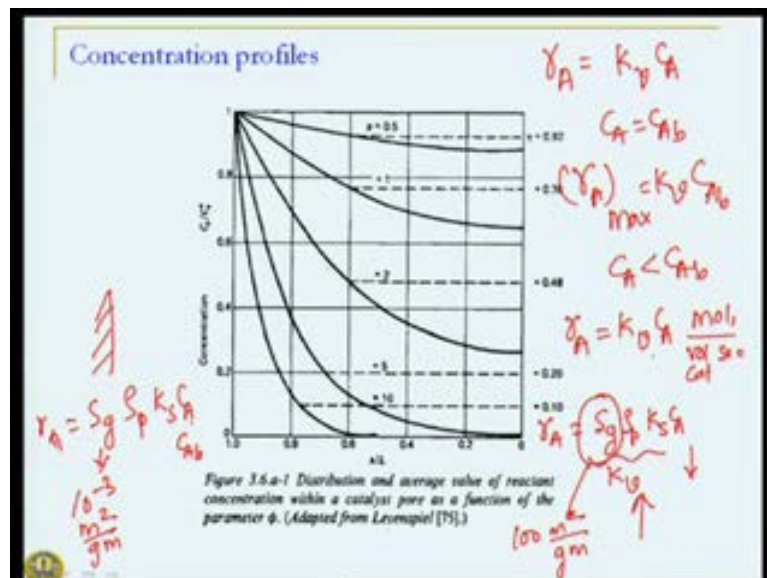
We see that concentration of reactant is more or less uniform and it is close to unity. That means, everywhere inside this catalyst pellet, there is a reactant and its concentration is same as the bulk concentration. Why is this happening? This is happening, because at low value of ϕ the diffusion time scales are much smaller compare to reaction that means diffusion is a fast process. So, the concentrations are close to unity. Whatever drop that you see is because some of that reactant is getting converted also to product. So, we expect some decrease, but it is more or less flat.

Compare to, if you look at the solutions with higher and higher value of, look at the solution for ϕ equal to 2, for example. There is a quite a steep gradient from outside surface to the inside of the catalyst pellet. With inside at the centre point concentration is only 30 percent that of outside. Of course, reaction is responsible for it partly, but also at higher value of ϕ , diffusion is a slow process. So, the gradient is much steeper. This can be further looked at when you look at the solution for ϕ Thiele modulus equal to 5, Thiele modulus equal to 10.

In fact, when Thiele modulus is 10 the reactant is pretty much consumed in upper 40 percent of the region that means, at value of ϕ the concentration drops down almost too

0 value in upper 40 percent of the region. The remaining 60 percent is there is no reactant, so there is obviously no reaction. Now, to summarize when we look at a plot like this, we see that diffusion creates gradients slower the diffusion rates higher is the higher is the gradient. Now, when the reaction rates are fast that is reaction rate is faster than the diffusion, we see lot of gradient. But diffusion is fast, the reaction concentrations are pretty much pretty much high. Now, that raises a question that, why should we then use porous catalyst? Because, the reaction would have been at its peak had the concentration be the bulk concentration.

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So, let's look at this situation once again. If you remember now, our rate of reaction is K_v into C_A . So since C_A is changing along the actual direction, the rate of reaction is also changing. And the rate of reaction is at its maximum, when C_A equals to C_{Ab} . So, r_{max} is K_v into C_{Ab} , but as we go inside the catalyst concentration is less than C_{Ab} , that is the diffusion effect. In fact, more so, if the diffusion is slow. If that is the case, then why have this catalyst in a porous form.

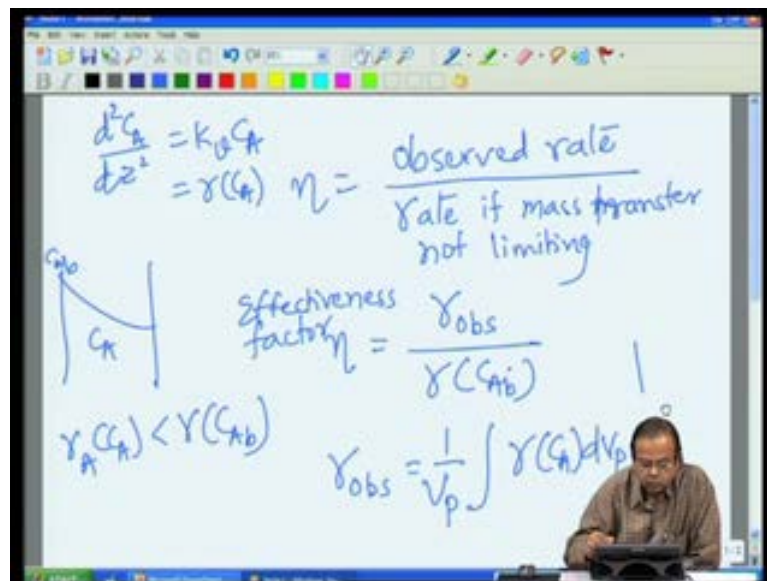
Why not have solid blocks? The reason for that is going back to our reaction, remember this is per, that is moles per volume of the catalyst, and let us say per second that is our reaction rate. Which essentially is, this is our intrinsic rate K_s which is moles per unit area multiplied by ρ of the catalyst pellet into the surface area. So, K_v is nothing but

this particular constant, this is concentration, this is K_s into c_a is moles per centimeter square or meter square per second, multiplied by S_g meter square per gram of the catalyst, multiplied by ρ_P which is gram per volume of the catalyst and that is how we get K_v .

Even though C_A is low when the catalyst is porous, K_e value is fairly high, because porous catalyst gives us as close as high as hundred meter square per gram of catalyst. So, whatever lost in reaction rate that occurs, because concentration is low is more than made up by surface area, which is high. This is my catalyst pellet, if I were to do the same reaction only on the surface everything else being same C_a may be C_{A_b} , but S_g will be of the order 10^3 meter square per gram. Compare this 10^3 to 10^{-3} , when the reaction occurs only on the surface that is catalyst is non-porous, v_{i_s} is where it is porous and reaction can occur inside.

And that is the reason, why we have porous catalyst, not solid block of catalyst. Now, since rate of reaction is changing along the actual direction the overall rate that you will get will be the average of this value. So, what will that rate be? Is one thing that we would like to calculate, but also we would like to calculate, how diffusion has increased or decreased the rate of reaction? For this particular kinetics, since concentrations are low and low concentrations means low reaction rate the overall rate would be lower compare to, if diffusion resistances were not there. So, let us try to bring in another concept to explore this idea further.

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So, let's go back to our mass balance equations. And say that our mass balance equation in the dimensionless form was k_b into C_A or let us just put it as, rate as a function of C_A . So now, because in my catalyst C_A is lower than the bulk concentration C_{Ab} , my rate of C_A is going to be less than the rate that I would get, if concentrations were C_{Ab} everywhere.

The question is how much lower? And to characterize this, let us bring in the concept of effectiveness factor, a new concept which says that there are two rates that I can have. One is the actual observed rate that I will get, because catalyst is poor. But then, I also have rate if mass transfer was not limiting that means, rate if the concentrations was C_{Ab} . So, we will put this in the more formal form, as saying that $r_{observed}$ divided by r of C_{Ab} . That is the observed rate divided by the rate, if concentrations were C_{Ab} . And the ratio of this we call it as effectiveness factor η . So, this is my concept of effectiveness factor, effectiveness factor η .

Now, how do I calculate these effectiveness factors? Now, what is my observed rate? Since my rate is changing all along the catalyst pellet. My observed rate is average. So, mathematically it is integral of r of C_A into along the volume or the volume of the pellet, average value. So, 1 over volume of the pellet so, this is my $r_{observed}$. Now, how do I calculate this $r_{observed}$?

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Handwritten equations on a whiteboard:

$$r_{obs} = \frac{dG_A}{dz^2} = K_p C_A = r(C_A) = \frac{1}{L} \int_0^L r(C_A) dz$$

$$D_{Ae} \left. \frac{dC_A}{dz} \right|_{z=L} - D_{Ae} \left. \frac{dC_A}{dz} \right|_{z=0} = \int_0^L r(C_A) dz$$

Diagram of a slab of length L with a concentration profile $C_A(z)$ and flux $D_{Ae} \frac{dC_A}{dz}$ at $z=L$.

$$\frac{1}{L} D_{Ae} \left. \frac{dC_A}{dz} \right|_{z=L} = \frac{1}{L} \int_0^L r(C_A) dz = r_{obs}$$

To calculate my r_{obs} , for this slab geometry I can also write this as taking the volume of the pellet that if this length is L and the cross sectional area that you see is A then this is nothing but 1 over A integral $r dC A dz$. That is I multiply and divide by area I get volume converted into the actual location, assuming the uniform cross sectional area. Now, let me once again write our mass balance equation. It is $d^2 C_A / dz^2 = D_{Ae}^{-1} K_p C_A$. So, $r_{obs} = r(C_A)$, as we wrote earlier.

Now, using the fact that I, if I take integral one time this will become $D_{Ae} dC_A / dz$. if I integrate this equation once at any z minus $D_{Ae} dC_A / dz$ at z equal to 0 , integrating this as integral r of $C A dz$. and what do we know about this at z equal to 0 , I am just putting it between two boundaries 0 and 0 and z or let us make it 0 and L . That is entire catalyst pellet, and then this will be between 0 and 0 and L .

Now, what do we know about this value? This is 0 or in other words, this R_{obs} r_{obs} is nothing but $D_{Ae} dC_A / dz$ at L , 1 over L will be 1 over L , integral 0 to L r of $D C A dz$ or r_{obs} . So, what does it say about r_{obs} ? If you look at these equations, see there is diffusion the flux at this point is $D_{Ae} dC_A / dz$ at z equal to L that is my flux. So, what this equation tells us is that, if you position, let us say yourself outside the or if the reactant is in the bulk. the way things are looked at from bulk, the average rate that is this observed rate, is actually the rate at which the material is

diffusing at this surface z equal to L . And of course, normalized by 1 over 1 over L . and that makes sense, because as far as this bulk is concern, there is no reaction in bulk. All it sees is that reactant is diffusing inside. So, the flux at which the reactant is diffusing given by this left hand side. This one over here must be same as the rate at which material is reacting on an average.

Because there is no accumulation, we are looking at steady state balance. So now, if we put all these things together, namely our definition of η .

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The image shows a whiteboard with handwritten mathematical derivations. At the top, it shows the differential equation for concentration C_A in a slab of thickness L :

$$D_{Ae} \frac{d^2 C_A}{dz^2} - D_{Ae} \frac{dC_A}{dz} \Big|_{z=0} = \int_0^L r(C_A) dz = r_{obs} L$$

Below this, it shows the integration of the differential equation to find the flux at $z=L$:

$$\frac{L}{L} D_{Ae} \frac{dC_A}{dz} \Big|_{z=L} = \frac{1}{L} \int_0^L r(C_A) dz = r_{obs}$$

Then, it defines the effectiveness factor η as the ratio of the observed rate to the rate based on the concentration at the surface C_{Ab} :

$$\eta = \frac{r_{obs}}{r(C_{Ab})} = \frac{1}{L} \cdot \frac{D_{Ae}}{k_m C_{Ab}} \frac{dC_A}{dz} \Big|_{z=L}$$

Finally, it shows the dimensionless form of the equation and the resulting expression for η :

$$\rightarrow \eta = \frac{1}{\phi^2} \frac{d\psi}{dz^*} \Big|_{z^*=1} = \frac{\tanh \phi}{\phi}$$

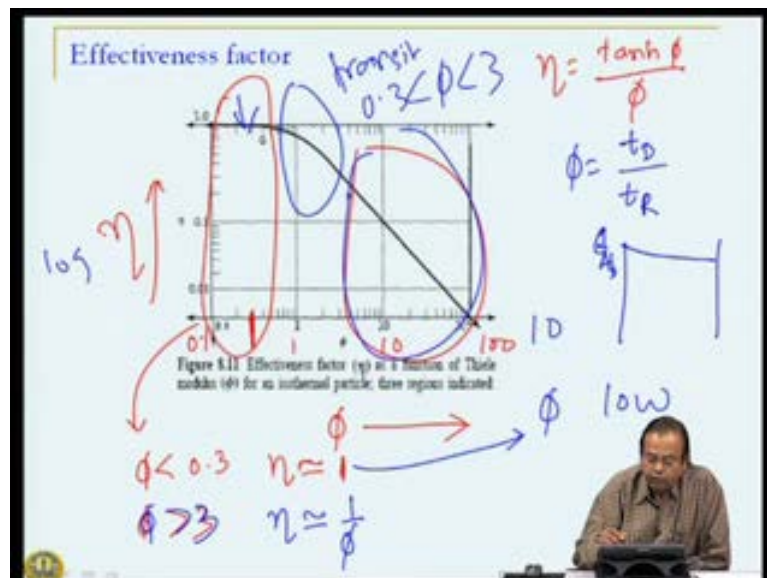
So, let us write down our η which is r observed by $r C A b$ is nothing but from this particular expression, is nothing but 1 over L into $D A e$ by $k m$ into $C A b$ or $k v$ into $C A b$ integral. Rather instead of integral, we are now writing it in terms of differential. So, that will be $D C a d z$ star $d z$ at z equal to L . And if you now make this equation dimensionless by using the same quantities, we end up with 1 over ϕ square $d u$ $d z$ star at z star equal to 1 . So, from going from this equation to this equation, all I have done is instead of $C A$ I have put u as $C A$ by $C A b$ and z star as z by z by L .

Now, it turns out that for linear system, you can actually solve these equations analytically. And you get a solution, $\tanh \phi$ by ϕ that is my effectiveness factor. Let us look at this equation more carefully and look at what is the how does this effectiveness

factor then depends on phi. To recap what we have done is, we have created a rating system for the observed rate by rating it with the rate, if rate of reaction if diffusion was not there.

And we called these rating factors as effectiveness factor. It turns out that for first order system in slab geometry, this effectiveness factor is given by $\frac{\tanh \phi}{\phi}$, where phi is a Thiele modulus $L \sqrt{\frac{D A e}{L^2 K v}}$ by $L \sqrt{\frac{D A e}{L^2 K v}}$. How does this solution look like?

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So, let us look at those solutions. And what we see here **what we see here is** an effectiveness factor on x-axis. So, this is my Thiele modulus **sorry** Thiele modulus on x-axis and effectiveness factor on y-axis. What we see here, this value is 0.1, 1, 10, 100 and so on. This is a solution of effectiveness factor being $\frac{\tanh \phi}{\phi}$. That is my solution that we see here.

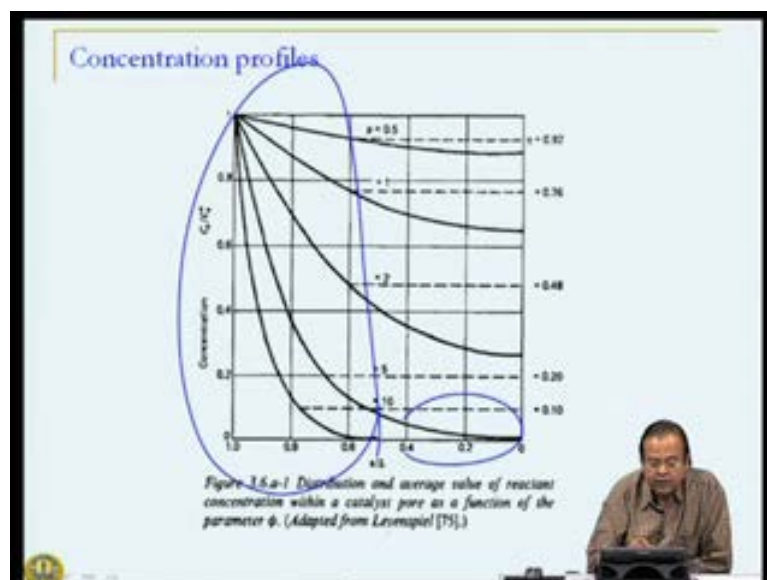
Now, we clearly see three different regimes. Regime one that is roughly about 0.3 up to 0.3 this particular regime, for phi less than 0.3 effectiveness factor is almost unity. For phi greater than three, another regime 1, 2, 3 over here in this regime, so this is this particular regime. This particular regime let me see, if I can change the **colour**, this particular regime that is phi greater than 3 **phi greater than 3**, what do we see? Remember

this is \ln plot.

So, this is \log plot and what we see is η is almost same as $1/\phi$. This is all mathematical and in between region is the transition region. That is $0.3 < \phi < 3$. This is all numerical values, but what does it physically mean. Remember, Thiele modulus is time of diffusion to time of reaction. So, ϕ value low that is this particular case implies that reaction rates are much higher than the diffusional time scales. Time scales of reaction are much higher than diffusional time scales.

That means, reaction is actually slower compare to diffusion. And if that is the case, then we saw in the earlier figure that concentration is flat more or less that is we are looking at concentration being close to unity. And therefore, it is not surprising to see that the effectiveness factor is close to unity in this particular region. What does it mean? That means, throughout the entire catalyst, since concentration is close to the bulk value, throughout this entire region, the reaction rate is more or less same. That is the observed rate is same as the rate, if reaction was to occur under bulk conditions and therefore, the rating parameter is 1.

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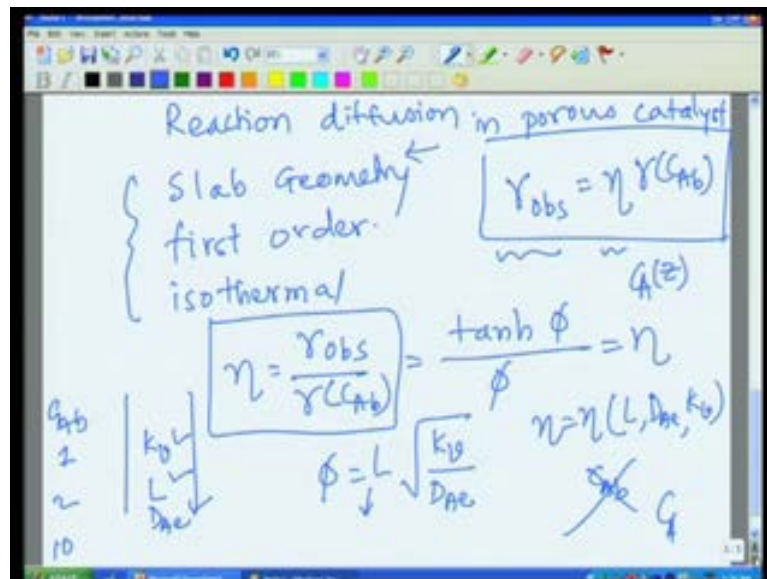


But if ϕ value is large ϕ value is large, let us say 5, 10, we see that there is very low concentration of reactant particularly in this particular region. Which means that the

observed rate that is the region that is contributing to reaction is only this region, if phi value is large. And here the concentrations are low that means, the average rate compare to that we obtained in the bulk now must be fairly low.

And in fact, that is what we see over here. And furthermore, when phi value increases, it exceeds the tan phi goes to unity. And therefore, eta becomes almost same as 1 over Thiele modulus phi and that is what we see in this particular region. So, this region is diffusion control, when Thiele modulus is large. This region is reaction control, when Thiele modulus is very small. And then we have a transition from Thiele modulus being between 0.3 and 3, where it is controlled neither exclusively by reaction nor by diffusion, but it is a region of mix control and hence we called it as transition period. Now, we said that.

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Let me recap what we have looked at so far. We considered reaction diffusion in porous catalyst porous catalyst; particularly the catalyst was with slab geometry. We considered first order reaction; we considered isothermal. Very nice ideal conditions for which you get even analytical solutions. Now, one of the things that we did, then what we saw is concentration as a function of distance in the catalyst pellet, but in terms of overall effect r observed by r at C A b is tan phi by phi Thiele modulus phi, where phi is L square root of K v by D A e. There are few implications of this result, before we go on to general

results which we follow.

First, let us look at this $\eta = \tan \phi / \phi$. So, what does this η depend upon and I mind you I am talking about only results only for these conditions mentioned over here at the moment, but they have far wider implications as we will see little later on. So, what do we get here? Effectiveness factor η depends on what? it depends on the catalyst length. So, it depends on effectiveness factor is a function of $L \sqrt{D A_e}$ and K_v .

These two L and $\sqrt{D A_e}$ are characteristic of the catalyst K_v is a reaction catalyst reaction characteristic. This of course is also characteristic of reaction, but it is more diffusion characterization and K_v is an indication of the reaction rate or kinetics characteristic of the catalyst. So, what it implies.

Nowhere in these equation for example, has $C_{A,b}$ occurred. It implies for this particular condition slab geometry first order isothermal, that the effectiveness factor does not depend on the bulk condition. So, whatever may be the concentration in the bulk that is, if I have my bulk concentration $C_{A,b}$ of some arbitrary unit 1, 2, 10. If the catalyst is same, that means, L is same, $\sqrt{D A_e}$ is same and the reaction is same, the effectiveness factor is same. it does not depend upon $C_{A,b}$, that is observation number one. The observation number two comes from this particular relationship. It says that r_{observed} is η into $r_{C_{A,b}}$ same equation.

Now, imagine you are doing the reaction with a porous catalyst. There is no way or it is very difficult, I should not say no way, because you never know what will happen in few years down the line. But by at large, there is no way that you can go inside the catalyst and measure the concentration of the reactant. All you can measure is the bulk concentration, but at the same time, you would also you would like to know what is the overall reaction or observed reaction rate. That is the average whatever is happening inside the catalyst pellet.

Now, if you look back that is r_{observed} is η into $r_{C_{A,b}}$, this tells us that to know r_{observed} . I know my bulk concentration and kinetics I need to know my η . And this information $\eta = \tan \phi / \phi$ tells us that η does not depend upon $C_{A,b}$ or for that matters $C_{A,b}$, but it depends on the characteristic of the catalyst. If I know what my catalyst is, I will know my observed rate of reaction by using these equations. Without

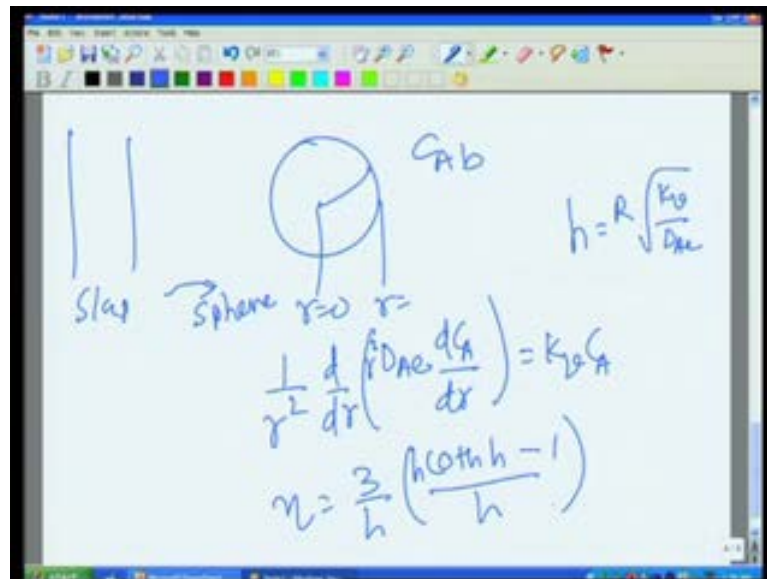
knowing, what is the concentration inside the catalyst pellet that is how is concentration C_A changing with z . And in my opinion, this is the most important result which is often use in design of catalytic reactor.

In design of catalytic reactor our dilemma is that we do not know what is there inside the catalyst. Yet we want to find out the observed rate. So, these relationship are observed is equal to η into r of C_A tells us, how to calculate observed rate without knowing, what is there inside the catalyst pellet. The only information i need to know is what is this effectiveness factor. Now for first order reaction, these effectiveness factors even does not depend upon the bulk condition. It depends only on the characteristic length of the catalyst, the diffusivity, the kinetic rate constant and so on.

Thus for a first order system, I know the complete information of the observed rate, if I know the characteristic of the catalyst. I do not have to know what are the reactant concentrations inside the catalyst. And this is one of the most important implications of this rating behavior. Why do I say it is most important? Because being engineers, we always try to approximate. If I know that this is the exact result under certain limiting condition. can I approximate my real situation to this limiting condition or how good or how far I am away from this condition, will tell me whether I could use the same approach or not. And let us try to look at such behavior.

By first relaxing our slab geometry concept and let us say that we have cylindrical geometry or spherical geometry.

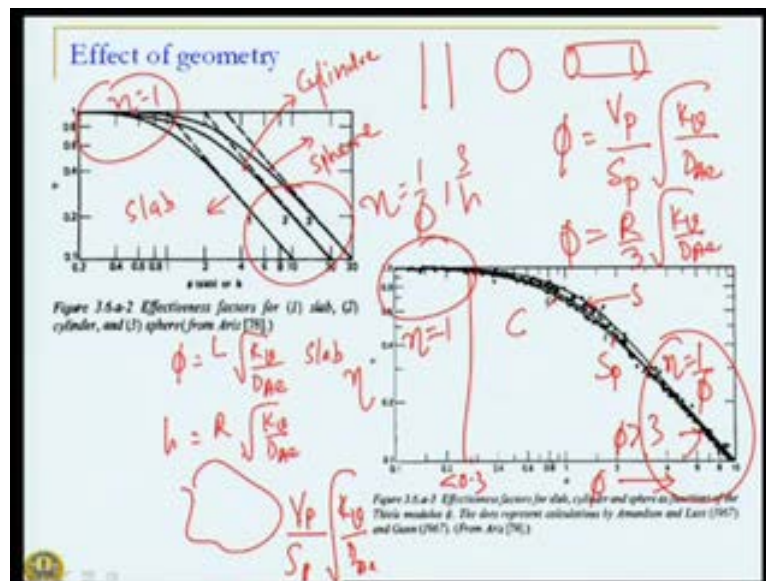
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My catalyst is now spherical like this. This is my r equal to 0, this is my r equal L , this is my bulk. So, my concentration will now change from the surface to the centre in this particular manner. Earlier, we considered slab. Now, slab geometry to sphere. So, I am relaxing my constrain on sphere. Now, if you do again the mass balance, we will get a mass balance, K_v into C_a . and then again this equation can also be analytically solve fortunately, and you can get effectiveness factor.

That is in this case for example, if you define the dimensionless constant H , as radius square root of D , square root of K_v by $D A_e$. We get effectiveness factor in this particular case, $\cot H$ minus 1 by H and if we now look at the solution of this particular scenario.

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What we are seeing here is solution for phi equal to 1 square root of K v by D A e or H equal to r by 3, this was for my slab geometry, this is for my. Let us say r square root of K v by D A e. So, that is my H, this is phi or h versus effectiveness factor. This result is for slab, this result is for sphere and this result is for a cylindrical geometry. Whether my catalyst is a slab or a sphere or a cylinder like this, remember various different shapes of catalyst. I get a result which qualitatively is very similar.

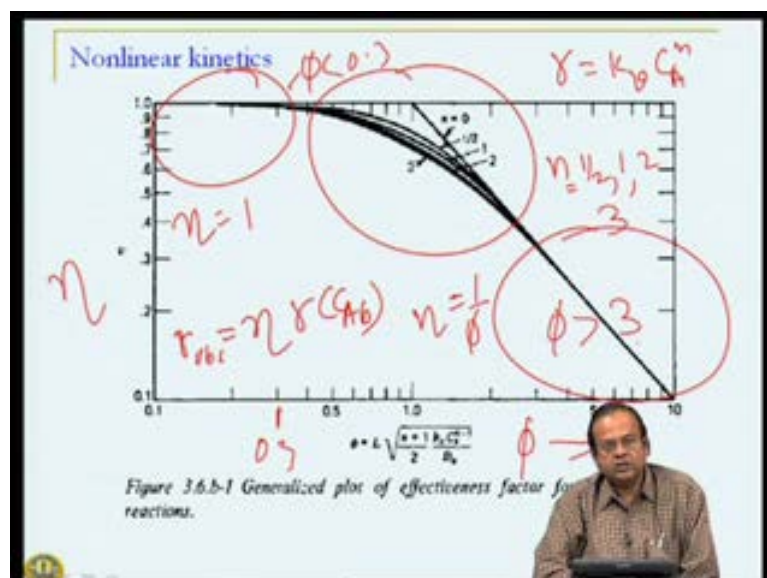
That is for some low value effect of this phi or h, it is unity, and eta is one. And here eta is 1 over phi, for example, for slab or 3 by H for sphere. So now, this is nice, because I can see use the same argument which we just few minutes back talked about for slab geometry. Equally applicable to the spherical geometry, but we can go a step further. For example, if we define our Thiele modulus phi, as volume of the pellet to the surface area of the pellet. And we are talking about only the external volume and surface area.

Now, divided by root K v by D A e D A e and plot this result phi as defined over here versus effectiveness factor. Remember this, for sphere what will it be? it will be 4 by 3 pi r cube divided by pi r square. So, this will be r by 3 square root K v by D A e. So, if we do that then we see that is different results for slab cylinder and sphere, they all collapse almost into a unique line.

You see here this is for cylinder, this middle one is for; the last one is for sphere the middle one is for, you cannot even probably see. I can see it, because I am sitting close to it. And this one is for this is for slab geometry; this is for sphere and for cylinder. And what are these dots that you see? Those dots are calculations done by Amundsen and loss go back in late sixties for any irregular shape. So, catalyst may be of this shape 3d shape. You define Thiele modulus, as volume of this catalyst pellet divided by the surface area the outside surface area square root K_v by $D A_e$. We get dots also falling onto almost same line. With a characteristic behavior η equal to 1 for Thiele modulus somewhere less than 0.3, η equal to $1/\phi$ for ϕ greater than 3 in this region. Or in other words, we almost have some kind of a unique relationship between η and ϕ , even though geometries are different, even though we have slab or sphere or cylinder or any irregular shape.

Then we see that the behavior in terms of η and ϕ is same. If that is a case, then for first order reaction to know observed rate. If I can approximate these behaviors by single line, I know my observed rate without knowing what is happening inside the catalyst pellet. This is as far as the first order reaction is concern, for any geometry. So, henceforth we will see our results mainly for slab geometry, because we know the similar things will come out for sphere, cylinder or irregular shape. And then relax what happens, when we have a first order kinetics.

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Instead of first order kinetics, we have nth order kinetics or in other words, let us look at these results. This is a result for effectiveness factor η versus Thiele modulus. We will talk about what is definition of Thiele modulus is in the next session. But before we conclude this session, let us look at this behavior. What we are seeing here is the result for power law kinetics $r = k C_a^n$. So, n is for example, half or 1 or 2 or 3 in this particular case. And we once again see amazing results, namely $\eta = 1$, when ϕ is less than 0.3, $\eta = 1/\phi$, when ϕ is greater than 3.

And intermediate regions where results are slightly different depending on the value of n , but the two extremes of reaction control and diffusion control, for any power law kinetics the effectiveness factor relationship with Thiele modulus is unique and this is a remarkable result. Because what it means is the definite equation of this line at least for limiting cases, I know r observed η as $r = k C_a^n$. But for general kinetics, I would not say that η is independent of bulk conditions or η which depends on Thiele modulus.

And Thiele modulus actually depends on the bulk conditions. How do we get that result, will be the topic of discussion. In the next session, followed by relaxing the constraint that the reaction is isothermal. So, we will consider non-isothermal reaction behavior, which we will do in the next session. Thank you